

Techno-economic assessment of polygeneration based on fluidized bed gasification

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ABSTRACT

The electric power sector contributes to about a quarter of the total CO₂ emissions worldwide. Therefore, in most mitigation scenarios for the climate change the share of low-carbon electricity supply increases from the current share of approximately 30% to more than 80% by 2050. A promising approach is the polygeneration concept for electrical power and chemicals based on integrated gasification combined cycle. It offers the possibility to capture CO₂ in a very efficient pre-combustion process and is able to accommodate the intermittent renewable power generation from wind and solar while operating the gasification island at full load by producing synthetic chemical products during times of low power demand. In this work, a process model of a polygeneration plant including a 350 MW_{el} combined cycle power plant with a fluidized bed dryer, a fluidized bed gasifier, a gas purification unit, a CO-Shift unit, a Rectisol acid gas removal, and a synthesis reactor array is developed. The model is used to investigate the specific CO₂ emissions and the process efficiency for different operation modes for power and methanol production. The model offers two options to introduce excess electricity from renewable sources into the process: via hydrogen from electrolysis or via additional drying and heating of the feedstock. The influence on the process efficiency and the economics are assessed.

1. Introduction

In 2015, more than 80% of the worldwide primary energy in 2015 came from fossil fuels leading to a high release of CO₂ to the atmosphere. One way to reduce the GHG-emissions is the further expansion of renewable energy sources, notably wind and solar power. In addition to low emissions, renewable energies have the advantage of negligible fuel costs. Nevertheless, they have the disadvantage of intermittent power generation and therefore cannot provide base load power. Furthermore, high flexibility is challenging for technological reasons, because load changes are a major cause of fatigue related component failures. In addition, highly flexible operation causes low capability factors and therefore reduces or even impedes profitability of these plants. Another promising approach to decarbonise the power generation field effectively and reduce the GHG-emissions of several industrial processes significantly is Carbon capture and Sequestration (CSS) [1,2].

Among the CCS techniques, pre-combustion together with an integrated gasification combined cycle power plant are particularly interesting. They allow separation of CO₂ along with other gases and impurities with well-established technologies. Furthermore, the efficiency of the power production is similar to or even higher than conventional coal fired power plants. Gräbner et al. [3] conducted a

thorough technical and economical comparison of entrained flow and fluidized bed gasification for hard coal and lignite for a pre-combustion IGCC approach. They found, that thermal efficiencies of 51.5% and 41.3% could be achieved with fluidized bed gasification without and with carbon capture, respectively. With fluidized bed gasification technologies in contrast to entrained flow gasifiers, a broader variety of difficult and low grade fuels can be used instead of or in addition to coal to increase feasibility even more [4]. Thus, several studies examined the co-gasification of coal or lignite with residues or biomass [5,6]. The technical feasibility of gasification of low grade fuels has been demonstrated in Värnamo, Sweden, in test operation over several thousand hours [7].

The general concept of poly- or co-generation of power and chemicals from coal is not a new approach. The advantage is an increased flexibility to react to market demands. Several examples and concepts can be found in the literature. Gao et al. [8] proposed a polygeneration system for the production of power and methanol based on coal gasification, similar to the one presented in this work. They investigated the steady-state operation with the aid of graphical exergy analysis. The results show that in a combined production the efficiency could be increased by 3.9% compared to separate production of power and chemicals. A different concept for steady-state operation was

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Abbreviations

AGR	Acid Gas Removal
ASPEN	Advanced System for Process Engineering
CCS	Carbon Capture and Sequestration
CFB	Circulating Fluidized Bed
CGE	Clean Gas Efficiency
EEX	European Energy Exchange
GHG	Green House Gases
HRSG	Heat Recovery Steam Generator
IGCC	Integrated Gasification Combined Cycle
IPCC	Intergovernmental Panel on Climate Change
LHV	Lower Heating Value
NGCC	Natural Gas Combined Cycle
SNG	Synthetic Natural Gas
$I_{CO_2,eq}$	Feedstock Specific Emission of CO ₂ Equivalents

$J_{CO_2,eq}$	Power Specific Emission of CO ₂ Equivalents
$K_{CO_2,eq}$	Product Specific Emission of CO ₂ Equivalents
\dot{m}	Mass Flow
p	Unitary Prices
P_{aux}	Power Consumption of Auxiliaries
P_{net}	Net Power Output
P_{tot}	Total Electrical Power Output
Pr	Profit
T_S	Saturation Temperature
T	Temperature
X	Mass Fraction
ΔH_R^0	Standard Enthalpy of Reaction
η_{CC}	Carbon Conversion
η_{cg}	Clean Gas Efficiency
η_{el}	Electrical Efficiency

considered by Guo et al. [9]. For power production, char-fired CFB boilers are used. The char is produced from lignite in pyrolyzers. The gases and tars released during pyrolyzation are separated. The tar is hydrogenated to synthetic crude oil. The coal gases are processed in a dry reforming process to obtain a syngas for production of methanol or other chemicals. The economic evaluation of this polygeneration process shows a significant higher internal rate of return of about 24% compared to a standalone power plant. Wolfersdorf et al. [10] looked investigated the annex concept to increase capability factors of coal fired power plants. In this approach, a small-capacity gasifier and a

subsequent gas cleaning and synthesis unit is linked to a larger power plant. At times of high electricity feed-in from renewables, energy from the power plant is used to produce hydrogen by electrolysis and for the synthesis process. This lowers the minimum operation of the power plant from 50% to around 33%. Further work with changes in the plant configuration has been done by Gootz et al. [11] and Forman et al. [12]. The polygeneration approach also has been envisioned for other fuels, e.g. for waste biomass by Gassner et al. [13]. Their approach utilizes hydrothermal gasification to produce power and SNG.

This paper aims at assessing the feasibility of the following four

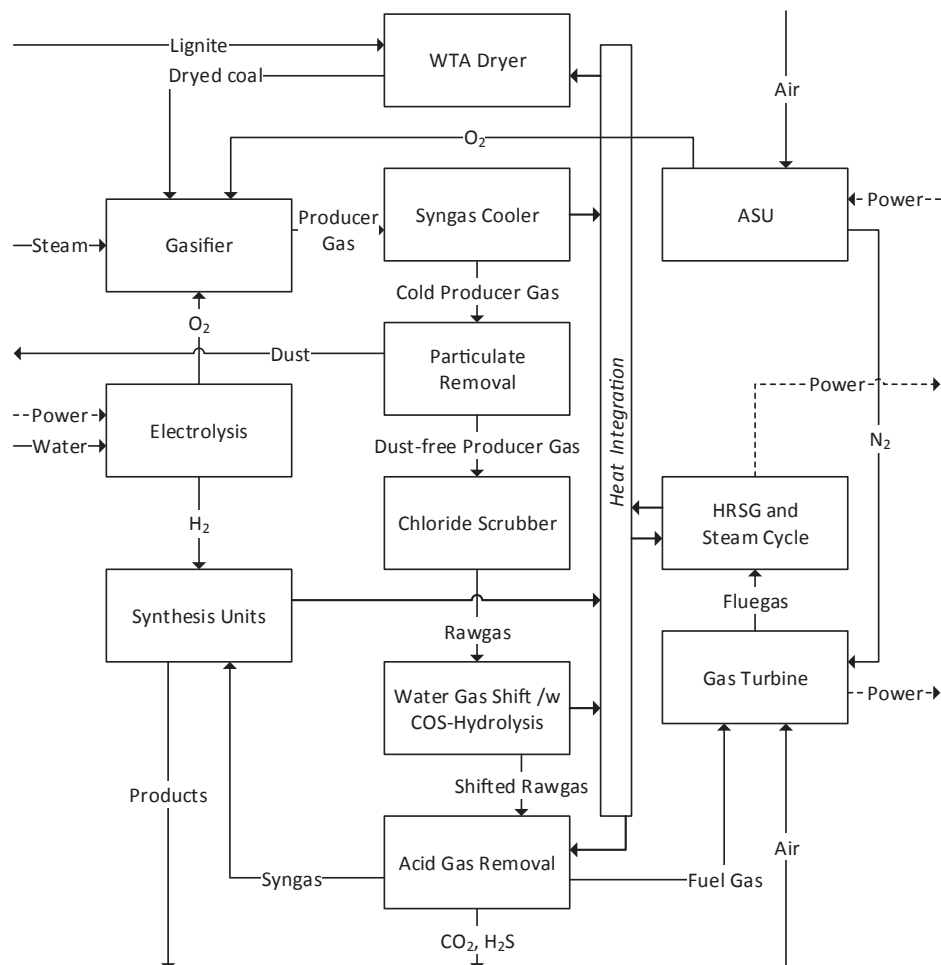


Fig. 1. Layout of the proposed polygeneration plant.

operation modes of a polygeneration plant centred around a fluidized bed gasification in the current market:

- Power production with an IGCC process without pre-combustion capture
- Power production with an IGCC process with pre-combustion capture
- Production of methanol from syngas with CO-shift operation
- Production of methanol from syngas with electrolyser operation

A steady-state process model of a commercial scale plant is implemented into Aspen Plus including all high-energy demanding process steps (i.e. pre-drying, gasification, gas cooling, WGS, AGR, synthesis, and combined cycle). This process model is used to simulate different operation modes. The results of the steady-state process simulation are then assessed with different economical, ecological and energetic key performance indicators. The economical assessment is conducted via the definition of inversion curves. These curves allow a statement on the favoured operation mode of an existing plant in different market situations, i.e. which mode would be used, if the plant is already built. The ecological assessment looks into the fate of greenhouse gasses. The energetic assessment presents the process efficiencies depending on the operation mode.

2. Process model

The process is depicted in Fig. 1. A raw low grade coal, Rhenish lignite, is introduced into a WTA dryer to efficiently reduce the moisture content [14]. The dried coal is sent to a HTW gasifier where steam and oxygen are added as gasification agents [15]. Oxygen is produced by an air separation unit and in some cases additionally by electrolysis of water. The produced syngas is treated in several processing steps, including syngas cooler, particulate removal, chloride scrubber, water gas shift unit and acid gas removal. The cleaned syngas then passes on to either a combined cycle power plant for power production or a synthesis unit for methanol synthesis. The steam produced in a heat recovery steam generator is used for power production in a steam cycle but also integrated in the other heat demanding and generating process steps for heat integration. For production of chemicals, additional hydrogen can be introduced into the synthesis unit from the electrolysis. Overall, the concept would produce chemical products (e.g. methanol), electric power, carbon dioxide, and H₂S from coal, air and excess electricity.

ASPEN Plus software was used to simulate the proposed process. This software was developed originally as a joint effort between the United States Department of Energy and Massachusetts Institute of Technology under the name of Advanced System for Process Engineering (ASPEN). The ASPEN software package is able to simulate steady-state processes with ASPEN Plus using flow sheet simulation.

The process model includes the WTA drying process, the HTW gasification, syngas cooling and scrubbing, WGS, AGR and the flexible production of chemicals and power in a synthesis plant and a combined cycle power plant respectively. The air separation unit and the water electrolysis are not modelled in the flowsheet but integrated with their specific energy demand.

The heat integration is done with an exergy balance. All heat streams above ambient temperature are rated with an exergetic efficiency of 59% which equals the exergetic efficiency of the reference steam cycle and is within typical values for steam cycles [16]. It is assumed that any additional heat stream can be used in the steam cycle with the same efficiency. For the cryogenic systems, an exergetic efficiency of 80% is assumed.

2.1. Model description

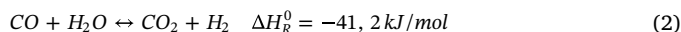
The WTA drying process is modelled with an equilibrium approach

and calculated depending on the coal moisture and the mass flow as well as the temperature of the steam. The relationship between the remaining moisture in the coal and the operation temperature of a fluidised dryer has been described by Buschsieweke [17]. For low operation pressures (~1.2 bar) that is expected in the proposed process the approximation equation is shown in Eq. (1) with T as the fluidised bed temperature and T_s as the saturation temperature. The heat is introduced into the dryer via a steam condenser. In this model it is assumed, that the temperature difference between temperature in the dryer and the steam from the steam cycle is 30 K.

$$X_{H_2O,DryCoal} = (T - T_s)^{-0.75} - 0.02 \quad (1)$$

The HTW gasification is modelled with an adiabatic Gibbs reactor operated at pressure of 30 bara. The carbon conversion is set to a typical value of 96% [18]. The gasification agent is composed of steam and oxygen. Oxygen is added to maintain a temperature of 1000 °C at the post gasification zone. It is taken from an ASU at 3.5 bar and 25 °C and compressed to the according pressure. The energy penalty for the uncompressed oxygen was set to 0.2 kWh/kg [19]. Alternatively, the oxygen is produced by water electrolysis. Here, the energy demand for the oxygen was set to 9 kWh/Nm³ [20]. The steam feed consists of high pressure steam with 30 bar and 350 °C. The flow rate is automatically optimized by the ASPEN Plus model for optimal syngas yield, i.e. highest cold gas efficiency in the model. The feedstock used in the simulation is based on lignite from the Rhenish area in Germany. The ultimate and proximate analysis of the actual fuel is shown in Table 1.

The syngas cooling and scrubbing are modelled by a heat exchanger and an adiabatic quench. The flow of quench water is set to yield a saturated syngas at a typical temperature of 150 °C [18]. The water gas shift unit is modelled with an equilibrium reactor for the shift reaction, Eq. (2). To yield the desired CO:H₂ ratio, there is a bypass around the water gas shift reactor. The separated streams are merged again after the reactor. The stoichiometric ratio for the methanol synthesis is set as stated in Eq. (3) [4].



$$\frac{(H_2 - CO_2)}{(CO + CO_2)} = 2.03 \quad (3)$$

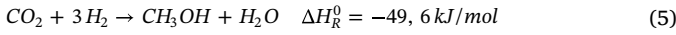
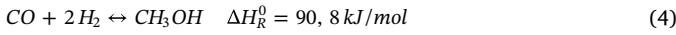
For the acid gas removal, a non-selective Rectisol process is implemented in ASPEN and is designed to be able to reach an H₂S content in the syngas of less than 0.1 ppm and a CO₂ content of about $3 \pm 0.5\%$. These values are recommended for methanol syntheses and sufficient for any combined cycle process [4]. The absorber is modelled with a 12-stage equilibrium model and the regeneration consist of a single flash, a four equilibrium stage desorber and a dewatering column.

For methanol synthesis, typical values from literature are used [4]. The syngas is compressed to the required pressure of 50 bar. The reaction is then modelled by an equilibrium reactor for the reactions shown in Eq. (4) and Eq. (5) and two flashes for product separation. The reactor is operated at 185 °C. Two flash reactors are used for product separation. The first flash separator is operated at 150 °C. The gaseous exhaust is recycled to increase the yield. The second flash is operated at ambient conditions.

Table 1

Proximate and ultimate analyses of the Rhenish lignite samples, oxygen calculated by difference.

Proximate analysis (wt.-%)				Ultimate analysis (wt.-%, daf)				
Water	Ash	Volatile Matter	Fixed Carbon	C	H	O (calculated)	N	S
54.0	9.3	20.1	16.6	70.1	4.84	23.12	0.75	1.19



The combined cycle is implemented with an validated model of the Prai Power Station NGCC heat recovery steam generator, described by Alobaid et al. [21]. The gas turbine was set to an air ratio of 1.2.

2.2. Performance indicators

The performance indicators have been chosen similar to the indicators defined by Boblenz et al. [22]. The energetic efficiency of the different process modes was explored using three main energetic indicators; the cold gas efficiency, the electrical efficiency and the carbon conversion. Hereby, the whole process chain depicted in Fig. 1 is considered, including all process steps shown.

The clean gas efficiency (η_{cg}) of any mode is defined according to Eq. (6) as the ratio of the heating value in the produced clean syngas to the heating value in the feedstock. It is derived from the definition of cold gas efficiency that is used for the characterization of gasifier performance. In Eq. (6) $\dot{m}_{Coal,mf}$ and \dot{m}_{Syngas} are the mass flow rate of the moisture free coal feed and the purified syngas, respectively, while LHV_{Coal} and LHV_{Syngas} are their respective low heating value.

$$\eta_{cg} = \frac{\dot{m}_{Syngas} LHV_{Syngas}}{\dot{m}_{Coal,mf} LHV_{Coal,mf}} \quad (6)$$

The electrical efficiency of the complete process (η_{el}) is defined in Eq. (7) as the ratio of the net power output (P_{net}) to the lower heat value of the introduced coal. In accordance with Eq. (8), the net power output (P_{net}) corresponds to the total produced electrical power (P_{tot}) after deducing the power consumption of auxiliary pieces of equipment (P_{aux}), which include all compressors, all pumps, all cryogenic systems, the ASU and the water electrolyser.

$$\eta_{el} = \frac{P_{net}}{\dot{m}_{Coal,mf} LHV_{Coal}} \quad (7)$$

$$P_{net} = P_{tot} - P_{aux} \quad (8)$$

The carbon conversion is defined as the ratio of the mass flow rate of reacted carbon from coal during gasification (\dot{m}_{rc}) to the mass flow rate of the carbon in the feedstock, Eq. (9). The carbon fraction X_C in the coal is set according to the Ultimate analysis.

$$\eta_{CC} = \frac{\dot{m}_{rc}}{\dot{m}_{Coal,daf} X_C} \quad (9)$$

For the environmental analysis, the feedstock specific emissions of carbon dioxide ($I_{CO_2,eq}$) are determined for the different operation

modes. This value is calculated by dividing the mass flow rate of the CO_2 emissions ($\dot{m}_{CO_2,eq}$) by the mass flow rate of the coal feed (\dot{m}_{Coal}). The CO_2 emission is defined as the sum of the different GHG emissions weighted by their respective Global Warming Potential in accordance with the IPCC definition.

$$I_{CO_2,eq} = \frac{\dot{m}_{CO_2,eq}}{\dot{m}_{Coal}} \quad (10)$$

In power generation modes, the power specific emission of carbon dioxide ($J_{CO_2,eq}$) are calculated. It corresponds to the mass flow rate of the CO_2 emissions ($\dot{m}_{CO_2,eq}$) related to the total generated electrical power, Eq. (11). On the other hand, the production specific emission of carbon dioxide ($K_{CO_2,eq}$) was determined for both operation modes of methanol synthesis. This is calculated by dividing the mass flow rate of the CO_2 emissions ($\dot{m}_{CO_2,eq}$) by the flow rate of product (\dot{m}_{prod}), Eq. (12).

$$J_{CO_2,eq} = \frac{\dot{m}_{CO_2,eq}}{P_{tot}} \quad (11)$$

$$K_{CO_2,eq} = \frac{\dot{m}_{CO_2,eq}}{\dot{m}_{prod}} \quad (12)$$

The economic assessment is realized by carrying out a profit comparison of the different operation modes by pairs. The profit (Pr) of every mode is calculated by deducing the variable operation costs from its revenues, Eq. (13). Here, $p_{Electricity}$, p_{prod} and $p_{CO_2,eq}$ are the unitary prices of electricity, chemical product and CO_2 allowances while \dot{m}_{prod} is the mass flow rates of produced methanol. This profit decides, whether at a given market scenario with set prices one operation mode will be favoured over another. In this approach, capital cost and fixed operating expenditures are disregarded, since they will be the same for a given plant, where only the mode of production has to be chosen. Hence, the result of the economical assessment will not give a statement about the leveled cost for electricity or methanol but will show, what operation regime would be chosen by an operator.

$$Pr = P_{net} \times p_{Electricity} + \dot{m}_{prod} \times p_{prod} - \dot{m}_{CO_2,eq} \times p_{CO_2,eq} \quad (13)$$

3. Results

In this chapter, the results of the energetic, ecological and economical assessment based on the steady-state model are presented.

3.1. Energetic and technical assessment

The clean gas efficiency as defined in Eq. (6) depends on the cold gas efficiency of the gasifier, which in turn depends on the moisture content of the gasifier feed stream as well as the correct feed rate of

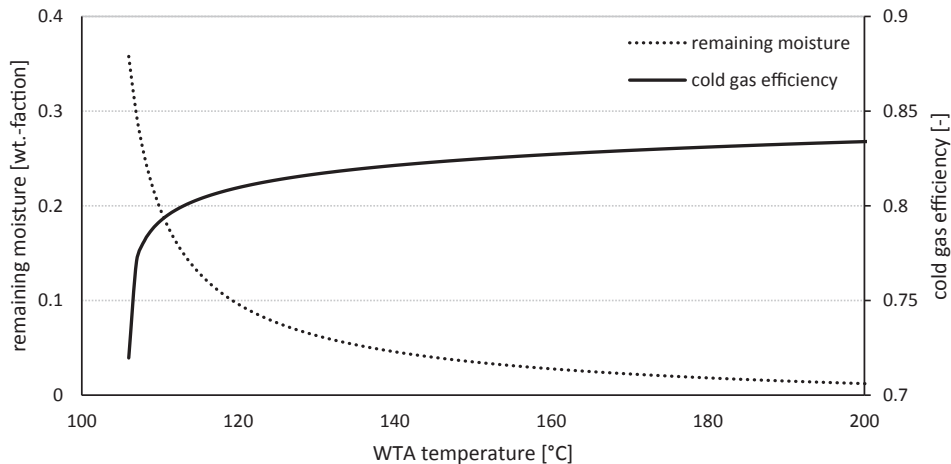


Fig. 2. Remaining moisture of the dried coal and the resulting cold gas efficiency of the gasifier for different temperatures in the WTA dryer.

oxygen and steam as gasifying agent. The feed rate of the oxygen and steam are endogenic in this model, as described in Section 2.1. The moisture content results from the WTA pre-drying, which in turn is controlled by the temperature of the WTA dryer. In Fig. 2 the remaining moisture in the coal and the cold gas efficiency is plotted over the WTA temperature. Generally, the remaining moisture decreases and therefore the cold gas efficiency increases with increasing temperature of the dryer. Further, the simulation results show, that above 115 °C in the dryer with a respective moisture content of around 16%, any further drying requires an according increase in steam feed for the gasifier. Hence, beyond the first increase of cold gas efficiency at low drying temperatures, the change can mostly be attributed to the lower energy demand of heating and evaporating the steam within the gasifier.

Nevertheless, the electrical efficiency of the complete process does not increase without limit, since the higher exergy of the steam for the gasifier has to be supplied by the syngas cooler or the steam cycle. The clean gas efficiency depends on the operation mode. The highest values are achieved around the aforementioned 16% remaining moisture and depicted in Fig. 3 for power production with and without carbon capture as well as the methanol production. The changes in clean gas efficiency are linked to the amount of CO and H₂O that are shifted to CO₂ and H₂ exothermally according to Eq. (3).

The electrical efficiency of the power production is shown in Fig. 4. The results for thermal efficiencies are about 3% lower than thermal efficiencies of IGCCs reported in literature for a lignite fired IGCC process without CCS [23]. This moderate divergence can be explained by divergences in the feedstock as well as an overestimation of power consumption of cryogenic devices in the simplified Rectisol AGR in the presented assessment. The energy penalty of around 4% for pre-combustion CCS is within the range that can be found in literature [23,24]. Nevertheless, some studies report significantly higher values of around 10% [3,25].

The performance of methanol synthesis is evaluated by determining the amount of carbon from the feedstock that is integrated into the product. The results are shown in Fig. 5 for the operation of 16% remaining moisture after coal drying for a stand-alone operation of the coal-to-methanol process as well as for a case with integration of renewable hydrogen from electrolysis. Hereby, the operation mode utilizing water electrolysis achieves 39.5% of carbon integration whereas the stand-alone approach exhibits a carbon integration of merely 24.7%. This considerable difference is associated with the necessary CO:H₂ ratio. Close to 50% of the producer gas carbon monoxide is converted to CO₂ during the water gas shift and withdrawn in the subsequent acid gas removal stage. When hydrogen from electrolysis is introduced into the synthesis reactor, the adjustment of the CO:H₂ ratio happens without varying the amount of CO in the producer gas.

3.2. Ecological assessment

One advantage of the presented polygeneration approach is the limited ecological impact compared to other uses of coal. This impact is indicated with an assessment of the GHG emissions and the fate of fossil carbon stored in the fuel, depicted in Fig. 6. During power production, without CCS, all carbon converted into syngas in the gasifier is released as CO₂. In power production mode with CCS, almost 90% of the fossil carbon is captured and taken from the plant as a pure CO₂ stream. The remaining CO₂ is emitted from the flue gas as well as different exhaust and waste streams.

In synthesis mode without additional hydrogen from water electrolysis, around 25% of the carbon is stored in the produced methanol. The emitted amount of carbon dioxide from the plant equals 211 g/kg coal, which is about double the amount that is emitted in power production mode with CCS. This is caused by the limited shift operation to reach the stoichiometric ratio as stated in Section 2.1.

In Fig. 7 the two methanol synthesis modes are compared. In both cases, the same amount of carbon is stored in the product and therefore

not emitted as direct CO₂ emissions. Without electrolysis, the emitted as well as the captured carbon dioxide is significantly higher than in electrolysis mode, which is directly connected to the different carbon conversion as described in Section 3.1.

3.3. Economical assessment

The economical assessment is conducted on the basis of inversion curves that describe the same price and profit ratios of different combinations of methanol, electricity and CO₂ allowance prices. As a comparison the prices from June 2018 at the EEX and NG-Tech [26] have been used. The price for CO₂ allowance was set at 15 €/t, for methanol at 408 €/t and for electricity at 40 €/MWh. This approach disregards financial, depreciation and other fixed and variable costs.

The first inversion curve in Fig. 8 compares the profit of power generation mode including carbon capture with that of methanol synthesis. Hence, the profits for electricity production and methanol production are normalized in regard to the CO₂ emission cost for each operation mode. Hereby, the methanol synthesis is slightly more advantageous according to the current prices situation, since the point of current prices ratios lies below the inversion curve. Above the inversion curve, the power generation becomes more beneficial. However, in periods of peak load it is likely that the electricity increases significantly. If the electricity price increases by more than 65%, the power generation becomes more favorable. This means, that the envisioned polygeneration plant would most likely work as a peak load power plant.

The second inversion curve in Fig. 9 compares the profit of power generation including carbon capture with that of power generation without carbon capture. Here, the power generation with carbon capture is more beneficial above the inversion curve. The results show that the IGCC power generation with carbon capture is more profitable according to the current price situation. This is caused by the comparably low energy penalty of around 0.5 MJ/kg for CO₂ absorption in the acid gas removal unit. This value is in the same order of magnitude as reported by [27].

The inversion curve shown in Fig. 10 balances the profit of methanol synthesis with water gas shift and that of methanol synthesis with water electrolysis. Hereby, the methanol synthesis with water gas shift is more beneficial according to the current price situation. This is led back to the relatively high energy demand of electrolysis and therefore high cost of electricity for this process. Nevertheless, the

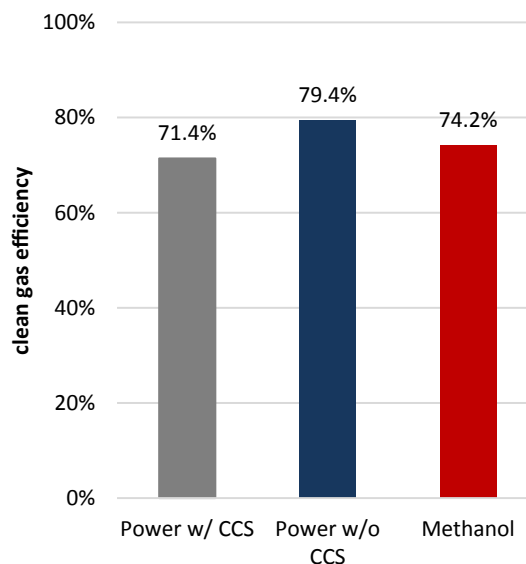


Fig. 3. Clean gas efficiency for different operation modes at 16% remaining moisture.

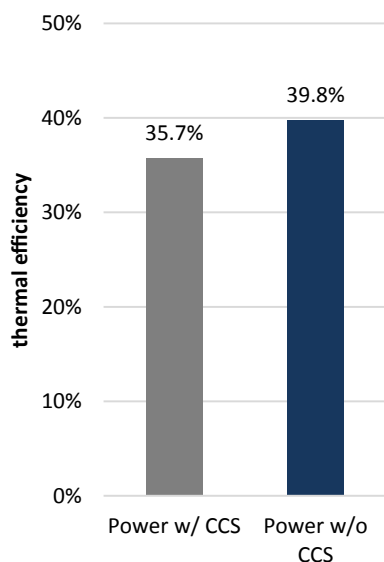


Fig. 4. Electrical efficiency for power production with and without carbon capture.

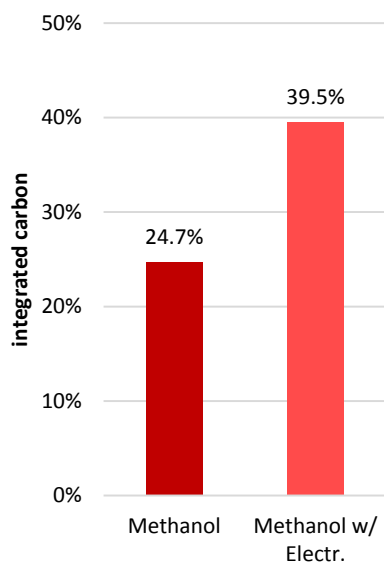


Fig. 5. Carbon from fuel integrated into product for the methanol synthesis for stand-alone operation and with integration of renewable hydrogen.

instantaneous electricity price is very volatile since it closely depends on the intermittent electricity production from the renewable power plants. The purchase of electricity for the adjustment of the CO:H₂ ratio via water electrolysis becomes more profitable from an electricity price's drop of about 47%. This corresponds to an industrial electricity price of round 21 €/MWh. This threshold was already approached several times in the off-peak periods according to EEX. Nevertheless, the feasibility of electrolysis in context of a polygeneration plant strongly depends on the additional investment cost for the electrolysis unit.

4. Conclusion and outlook

The presented work aimed at demonstrating a novel polygeneration concept for the highly flexible production of chemicals and electricity

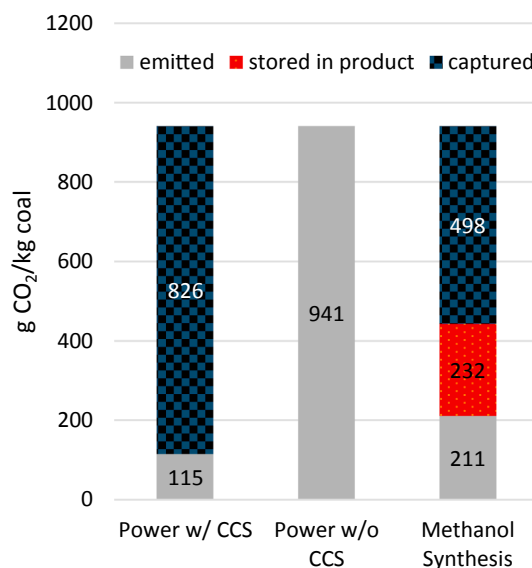


Fig. 6. Fate of carbon of the coal feed for different operation modes.

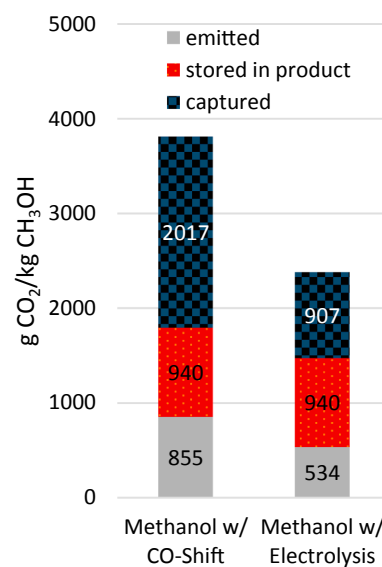


Fig. 7. Product specific emissions for methanol synthesis with ratio adjustment via CO-shift or electrolysis.

with steady-state process simulation. To achieve this, an extensive model has been implemented in ASPEN Plus and several operation modes have been assessed using different performance indicators.

The results show, that the envisioned polygeneration power plant would fit into the current energy and methanol market as a peak load power plant. During times of low energy demand, the plant would produce methanol from coal. The current prices for GHG emission allowances would promote an operation mode without carbon capture. Since a potent acid gas removal has to be integrated into the plant, the marginal cost for carbon capture is still quite low and could become feasible in the future. An integration of electrolysis is not economically feasible under current circumstances. Even though the electricity prices decrease below the threshold quite often where the marginal cost would favor a production of hydrogen from electrolysis, the typically high investment cost for electrolysis units would prohibit such a plant configuration.

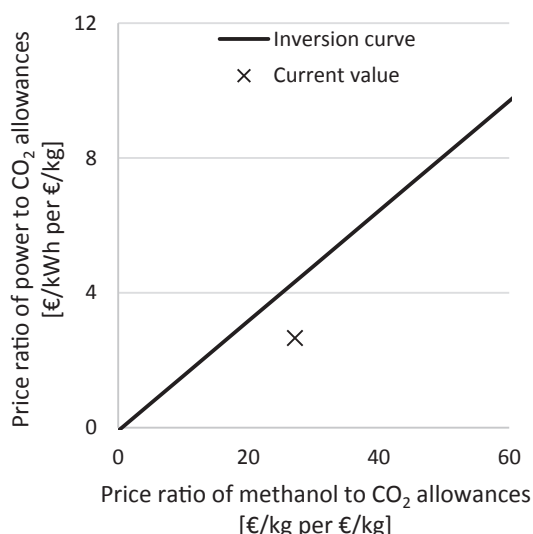


Fig. 8. Inversion curve for the production decision between methanol and power.

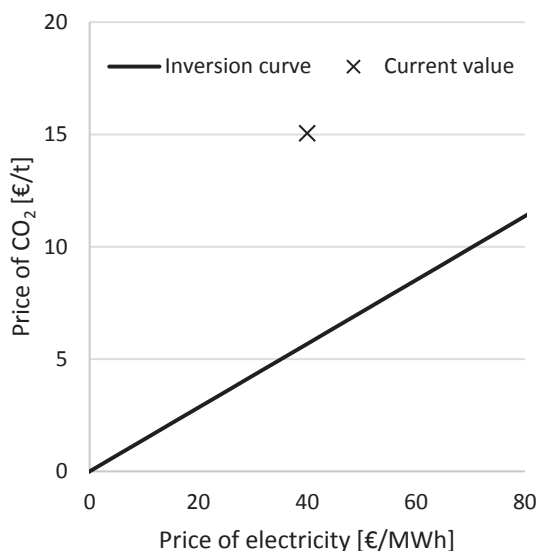


Fig. 9. Inversion curve for the production decision between power with and without carbon capture.

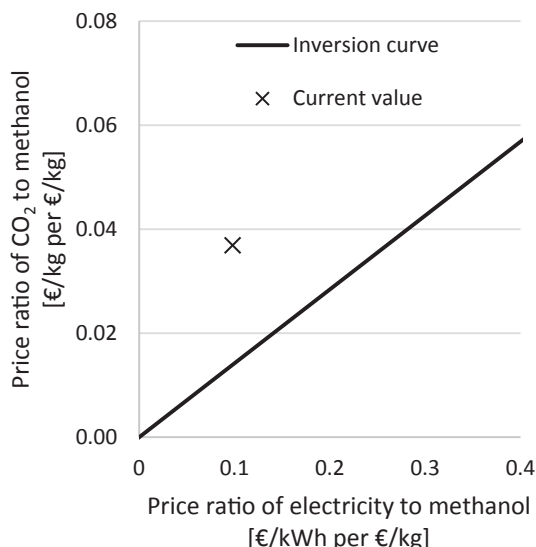


Fig. 10. Inversion curve for the production decision between methanol with or without hydrogen from electrolysis.

Based on this work, an analysis of the investment cost and a calculation of the levelized cost of electricity will be conducted in the future. Further, possible chemical products will be included into the model.

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